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Mixed-parity superconductivity in Sr_2RuO_4

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We show that in Sr_2RuO_4 the Fermi surface geometry as inferred from angle resolved photoemission experiments has important implications for a pairing interaction dominated by incommensurate, strongly anisotropic, spin fluctuations. For a spin fluctuation spectrum consistent with inelastic neutron-scattering experiments the system is close to an accidental degeneracy between even parity spin singlet and odd parity spin triplet channels. This opens the possibility of a mixed parity order parameter state in Sr_2RuO_4. We determine the stable and metastable order parameter phases at low-temperatures and discuss especially phases with order parameter nodes.

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The study of the superconducting material Sr_2RuO_4,[1] has attracted considerable experimental and theoretical attention because of its peculiar low energy electronic properties.[2] It is a rare example of a multiband superconductor with possibly spin triplet pairing symmetry.[3] Based on muon spin relaxation measurements[4] an early suggestion for the order parameter symmetry was the time-reversal symmetry breaking "$p_x \pm i p_y$"-state.[2] Recent experiments,[5–7] on the contrary, favor order parameters with nodes.[8] The spin excitation spectrum in Sr_2RuO_4 as studied by inelastic neutron scattering (INS) reveals spin fluctuations in a crossover regime between ferromagnetic and antiferromagnetic fluctuations. The dominant contributions to the spin susceptibility, $\chi(\boldsymbol{q})$, are located around $\boldsymbol{Q}=(1\pm\delta,1\pm\delta,0)\pi/a$, with an incommensurability $\delta\approx 0.4$.[9] Similar results have also been suggested theoretically finding $\delta\approx 1/3$.[10] It was suggested that in the crossover regime between ferromagnetic and antiferromagnetic spin fluctuations the order parameter changes from *p*-wave to *d*-wave.[10,11] As experimental results become more reliable, it becomes possible to test if a pairing mechanism via incommensurate spin-fluctuations is consistent with (a) the spin fluctuation spectrum as measured in INS, (b) the electronic structure as tested by angle resolved photoemission and de Haas-van Alphen spectroscopy, and (c) the experimental implications for the order parameter symmetry.

In this paper we study the possible pairing symmetries for the order parameter in Sr_2RuO_4 resulting from a pairing interaction dominated by incommensurate, strongly anisotropic, spin fluctuations. Given the measured Fermi surface geometry in Sr_2RuO_4,[12,13] we find that the system is close to two accidental degeneracies between the triplet E_{2u} superconducting state and one of the singlet superconducting states either in the B_{1g} (predominantly *d*-wave) channel or in the A_{1g} (predominantly *g*-wave) channel. This opens the possibility of a superconducting state with mixed parity order parameter in Sr_2RuO_4. The possibility of such superconducting states was discussed some time ago in connection with UPt_3.[14] We also study the different possible stable and metastable low-temperature phases in Sr_2RuO_4 for the three cases that either of the three relevant order parameter symmetries E_{2u}, B_{1g}, or A_{1g} is dominant. We find both nodeless solutions and order parameters with nodes. A dominant triplet order parameter is supported only in a surprisingly small incommensurability region near the experimentally determined value $\delta\approx 0.4$.

NMR experiments suggest a strongly anisotropic spin susceptibility, with only the χ_{zz} peaked around the incommensurate wave vector $\boldsymbol{Q}$.[15] Thus, we consider an anisotropic model with $\chi^z \equiv \chi_{zz} \gg \chi_{xx}=\chi_{yy}\equiv\chi^{\perp}$ and neglect off-diagonal components ($\chi_{xy}=0$). For χ^z we assume the following form ($a=1$ in our units)

$$\chi^z(\boldsymbol{q})=\sum_{\delta_{x,y}=\pm\delta}\frac{\chi_Q/4}{1+4\xi_{\rm sfl}^2\left(\cos^2\frac{q_x-\delta_x}{2}+\cos^2\frac{q_y-\delta_y}{2}\right)}. \tag{1}$$

This model susceptibility has three parameters. The overall magnitude, χ_Q, determines the coupling strength in the dominant pairing channel, and thus the superconducting transition temperature T_{c0}. The other two parameters, the spin-spin correlation length, $\xi_{\rm sfl}$, and the degree of incommensuration, δ, determine the relative coupling strengths in the different symmetry channels, defining the symmetry of dominant and subdominant components. The specific form of the susceptibility allows a smooth crossover from antiferromagnetic fluctuations, $\boldsymbol{Q}_{AF}=(1,1,0)\pi/a$, to ferromagnetic, $\boldsymbol{Q}_{FM}=(0,0,0)$, by tuning δ from 0 to 1. Extracting the values of $\xi_{\rm sfl}$ and δ from the INS data[9] gives $\xi_{\rm sfl}\approx 4.0a$ and $\delta\approx 0.4$. The effective pairing interaction via spin fluctuation exchange is determined by the coupling function $g(\boldsymbol{p})\chi^i(\boldsymbol{p}-\boldsymbol{p}')g(\boldsymbol{p}')$. The coupling between spin fluctuations and quasiparticles, $g(\boldsymbol{p})$, is approximated in what follows by a constant, g. To reproduce the structure of the experimentally probed three-sheet Fermi surface[12,13] we use the tight-binding dispersions

$$\epsilon_{\boldsymbol{p}}^i=2t_x^i\cos p_x+2t_y^i\cos p_y-4t'^{,i}\cos p_x\cos p_y-\mu^i. \tag{2}$$

The band index i labels the xy, xz, and yz bands. The parameters of the dispersions $(t_x^i, t_y^i, t'^{,i}, \mu^i)$ are taken from Ref. 16. Additionally, a hybridization of the xz and the yz bands is given by $t_\perp = 0.1$ eV.[17]

The order parameter in weak coupling BCS theory is determined by the well-known nonlinear BCS self-consistency equation. The smallness of the gap in Sr_2RuO_4 of about 1 meV,[18] allows us to restrict momentum summations in this gap equation to the vicinity of the Fermi surface. In a standard way this procedure leads to a replacement of the momentum sum by a Fermi surface average, $\langle \cdots \rangle_{\boldsymbol{p}_f}$, weighted with the angle resolved density of states, $N(\boldsymbol{p}_f) = N_f n(\boldsymbol{p}_f) = N_f |\boldsymbol{v}_f(\boldsymbol{p}_f)|^{-1} / \langle |\boldsymbol{v}_f(\boldsymbol{p}_f')|^{-1} \rangle_{\boldsymbol{p}_f'}$. N_f is the total density of states at the Fermi level and $\boldsymbol{v}_f$ is the Fermi velocity. We obtain $N_f = 1.78$ states/eV per spin and unit cell. Introducing the dimensionless coupling function $\overline{\chi} = N_f g^2 \chi$, the weak coupling gap equation reads

$$\Delta_{\alpha\beta}(\boldsymbol{p}_f) = - \sum_{i=\{x,y,z\}} \sum_{\gamma\delta} \langle \sigma^i_{\alpha\gamma} \overline{\chi^i}(\boldsymbol{p}_f - \boldsymbol{p}_f') \times \sigma^i_{\beta\delta} n(\boldsymbol{p}_f') f_{\gamma\delta}(\boldsymbol{p}_f') \rangle_{\boldsymbol{p}_f'}, \tag{3}$$

where $f_{\gamma\delta}(\boldsymbol{p}_f) = T \Sigma_{\epsilon_n}^{\epsilon_c} \int d\xi_p F_{\gamma\delta}(\boldsymbol{p}_f, \epsilon_n; \xi_p)$, F is the anomalous propagator, ϵ_n fermionic Matsubara frequencies, and ϵ_c is the usual frequency cutoff. The anisotropic interaction in Eq. (3) breaks spin rotational symmetry, but since each $\boldsymbol{p}_f$ state is doubly Kramers-degenerate in zero field, we can still decompose $\Delta_{\alpha\beta}$ and $F_{\gamma\delta}$ into (pseudo-)spin singlet (s) and (pseudo-)spin triplet (t) components[19] and arrive, for $x = s, t$, at

$$\Delta_x(\boldsymbol{p}_f) = - \langle V_x(\boldsymbol{p}_f - \boldsymbol{p}_f') n(\boldsymbol{p}_f') f_x(\boldsymbol{p}_f') \rangle_{\boldsymbol{p}_f'} \tag{4}$$

with $V_s = \overline{\chi^\perp} + \overline{\chi^z}/2$, $V_t^z = -\overline{\chi^\perp} + \overline{\chi^z}/2$, $V_t^\perp = -\overline{\chi^z}/2$. Isotropic spin fluctuations ($\chi^\perp = \chi^z$) support triplet superconductivity only for nearly ferromagnetic enhancement. In the case of extreme anisotropy the coupling functions for singlet and triplet pairing with the $\boldsymbol{d}$ vector in the $\hat{z}$ direction have equal sign and magnitude, $V_s = V_t^z = \overline{\chi^z}/2$. In addition, a second triplet-pairing state with $\boldsymbol{d} \perp \hat{z}$ is possible. It couples via $V_t^\perp = -\overline{\chi^z}/2$.

In order to study the symmetry of the superconducting state near T_{c0} we determine numerically the eigenvalue spectrum of the integral kernel in Eq. (4), following Ref. 20. The resulting complete set of orthogonal basis functions $\mathcal{Y}_\mu^\Gamma(\boldsymbol{p}_f)$ can be classified according to the irreducible representations (Γ) of the crystal group D_{4h}. The corresponding eigenvalues, λ_μ^Γ, determine the coupling constants for the μth basis function in the symmetry channel (Γ).[21] The most attractive (negative) eigenvalue in representation (Γ), $\lambda^\Gamma = \min_\mu(\lambda_\mu^\Gamma)$ is eliminated in favor of a transition temperature for order parameter symmetry (Γ) in the usual way, $T_{c,\Gamma} = 1.13 \epsilon_c \exp(-1/|\lambda^\Gamma|)$.

The dominant coupling constant λ^{Γ_0} determines the superconducting transition temperature T_{c0} and the symmetry (Γ_0) of the superconducting phase near T_{c0}. Once an order parameter component in a certain representation (Γ) nucleates, the other components $\mu(\Gamma)$ in the same representation are usually induced by the presence of the first component. Thus, physical transitions between different superconducting phases only occur when additional symmetries are spontaneously broken. Such subdominant transitions are suppressed below the value $T_{c,\Gamma}$ in the presence of a dominant order parameter. In Fig. 1 we study which of the different possible phases nucleates first at given $\xi_{\rm sfl}$ and δ and determine points of accidental degeneracy between two different order parameter phases as a function of these parameters. We show the dependence of the attractive eigenvalues on δ for $\chi^\perp = 0$, fixing $\xi_{\rm sfl}$ at $4.0a$, a value which should closely correspond to the actual value in Sr_2RuO_4.[9] The relevant phases near T_{c0} are the even-parity one-dimensional irreducible representations A_{1g} and B_{1g}, which give spin-singlet superconductivity, and the odd-parity two-dimensional irreducible representation, E_{2u}, rendering a spin-triplet channel for superconductivity. At small values of incommensuration, up to $\delta \approx 0.35$, the system prefers the B_{1g}-pairing channel. Above this point there is a region, $0.35 \lesssim \delta \lesssim 0.42$, of spin-triplet pairing in the E_{2u} channel with $\boldsymbol{d} \| \hat{z}$. Beyond $\delta \approx 0.42$, the pairing state is spin-singlet with A_{1g} symmetry, bounded by a narrow region of B_{1g} pairing starting around $\delta \approx 0.61$. In the large-δ range there is again triplet pairing, but now with $\boldsymbol{d} \perp \hat{z}$. The two accidental degeneracy points of interest for us are $B_{1g} \oplus E_{2u}$ for an incommensuration $\delta \approx 0.35$ and $A_{1g} \oplus E_{2u}$ for $\delta \approx 0.42$. Both points are remarkably close to the experimental value of $\delta \approx 0.4$ and the theoretically predicted value of $\delta \approx 0.33$. Calculations with an additional component $\chi^\perp$ resulted into accidental degeneracies even closer to each other. We also performed calculations for $\delta \approx 0.4$ and varying $\xi_{\rm sfl}$ showing that the presence of the accidental degeneracies near $\delta = 0.4$ is a robust feature for $\xi_{\rm sfl} > 2a$.

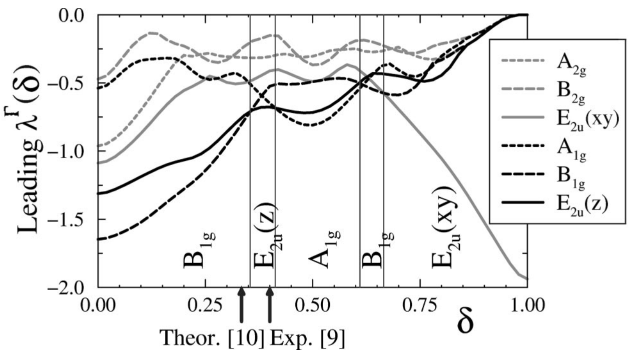

FIG. 1. The most attractive eigenvalues, λ^Γ, for the different irreducible D_{4h} crystal symmetry group representations, (Γ), as a function of the magnetic incommensuration parameter δ at $\xi_{\rm sfl} = 4a$. Even-parity representations are A_{1g}, A_{2g}, B_{1g}, and B_{2g}. The odd-parity states have either $\boldsymbol{d} \| \hat{z}$ $[E_{2u}(z)]$ or $\boldsymbol{d} \perp \hat{z}$ $[E_{2u}(xy)]$. The regions, separated by vertical lines, are marked by the representation of highest T_c.

Next we study the possible low temperature superconducting phases close to the accidental degeneracies. We concentrate on the three cases where either of the three symmetry channels is slightly dominant. We chose $\delta = 0.3$ for a dominant B_{1g} channel, $\delta = 0.4$ for a dominant E_{2u} channel,

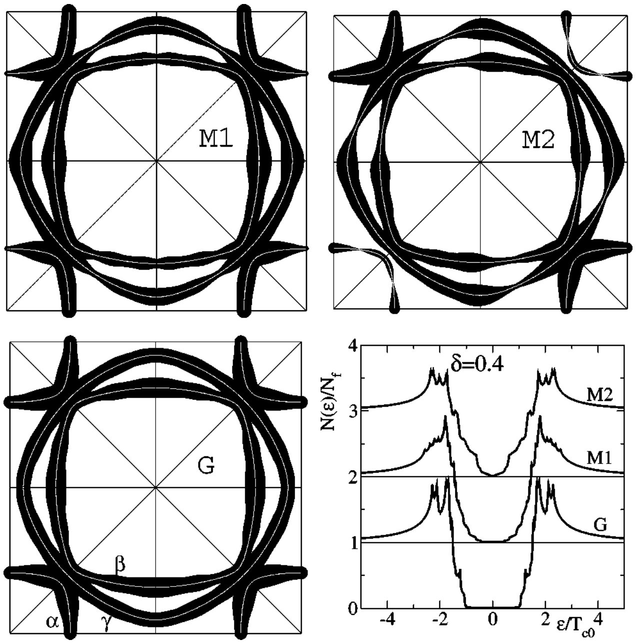

FIG. 2. The magnitude of the order parameter, $|\Delta_+(\boldsymbol{p}_f)|$, at zero temperature for the superconducting ground state (G) and the two metastable states ($M1$,$M2$), calculated for $\delta=0.4$ and $\xi_{\rm sfl}=4a$. The three Fermi sheets (α, β, and γ) are marked by thin white lines and $|\Delta_+(\boldsymbol{p}_f)|$ by the thickness of the black lines. The ground state is of pure E_{2u} symmetry, the metastable states mix E_{2u}, A_{1g}, and B_{1g}, resulting in mixed parity states. The corresponding total density of states, $N(\epsilon)$, for each state is shown in the lower right panel.

and $\delta=0.45$ for a dominant A_{1g} channel. To determine the superconducting state at low temperatures we solve the nonlinear gap equation, Eq. (4). Expanding the order parameter with respect to the set of basis functions $\mathcal{Y}_\mu^\Gamma(\boldsymbol{p}_f)$ we obtain the order parameter components Δ_μ^Γ for each representation. In the case of a mixed parity state a mixture of even parity, $\Delta^{(e)}(\boldsymbol{p}_f)$, and odd parity, $\Delta^{(o)}(\boldsymbol{p}_f)$, basis functions occurs as

$$\Delta_\pm(\boldsymbol{p}_f)=\sum_\mu \Delta_\mu^{(e)}\mathcal{Y}_\mu^{(e)}(\boldsymbol{p}_f)\pm\sum_\mu \Delta_\mu^{(o)}\mathcal{Y}_\mu^{(o)}(\boldsymbol{p}_f). \tag{5}$$

Once parity is broken, each of the doublets, $\psi_\uparrow\psi_\downarrow$ and $\psi_\downarrow\psi_\uparrow$, acquire separate order parameters, $\psi_\uparrow\psi_\downarrow$ with $\Delta_+(\boldsymbol{p}_f)$ and $\psi_\downarrow\psi_\uparrow$ with $\Delta_-(\boldsymbol{p}_f)$, and $\Delta(\boldsymbol{p}_f)$ has the required antisymmetry since $\Delta_\pm(\boldsymbol{p}_f)=-\Delta_\mp(-\boldsymbol{p}_f)$. As there may be several possible superconducting states, each state being a local minima in the free-energy, we compute the free energy of each candidate state using the Serene-Rainer free-energy functional,[22] and select the state of lowest energy as the low-temperature phase. As the temperature evolution of the order parameter depends on $T_{c,\Gamma}/T_{c0}=\exp(1/\lambda^\Gamma-1/\lambda^{\Gamma_0})$, it is dependent on the value of the most negative eigenvalue λ^{Γ_0}, which is determined by the parameter $g^2\chi_Q$. We have chosen $g^2\chi_Q/(2\pi)^2=1$, leading to eigenvalues $\lambda^\Gamma\lesssim 1$, appropriate for weak coupling.

In Fig. 2 we plot the magnitude of the superconducting gap as a function of position on the Fermi surface, $|\Delta_+(\boldsymbol{p}_f)|$, that minimizes the free energy at $\delta=0.4$ and with $\xi_{\rm sfl}=4a$. We find three local minima: one ground state (G) and two metastable states ($M1$ and $M2$). The free energy difference at zero temperature between $M1$ and G corresponds to only 9% of the ground-state condensation energy (it amounts to 22% for $M2$). G is of pure E_{2u} symmetry. $M1$ is symmetric around the p_x and p_y axis and has points with small gap values on the α sheet. $M2$ is symmetric around the diagonals $p_x\pm p_y$ and has nodes on the α sheet. Also shown in Fig. 2 is the total (angle-averaged) density of states (DOS) at the Fermi surface. The DOS is fully gapped for the ground state. The first metastable state shows a DOS with a much smaller excitation gap, and the DOS for the second metastable state shows low-energy nodal excitations originating from the α sheet. The metastable states break spin-rotation symmetry and parity. The amplitudes $\Delta_\mu^{(e,o)}$ have a zero relative phase for basis functions within the same parity, and a relative phase difference of $\pm\pi/2$ for basis functions with opposite parity; thus, $\Delta_-(\boldsymbol{p}_f)=\Delta_+(\boldsymbol{p}_f)^*$. In the ground state the odd-parity basis functions with same eigenvalue have a $\pi/2$ relative phase difference, giving a "$p_x\pm ip_y$" state. We emphasize that all three solutions break time-reversal symmetry.

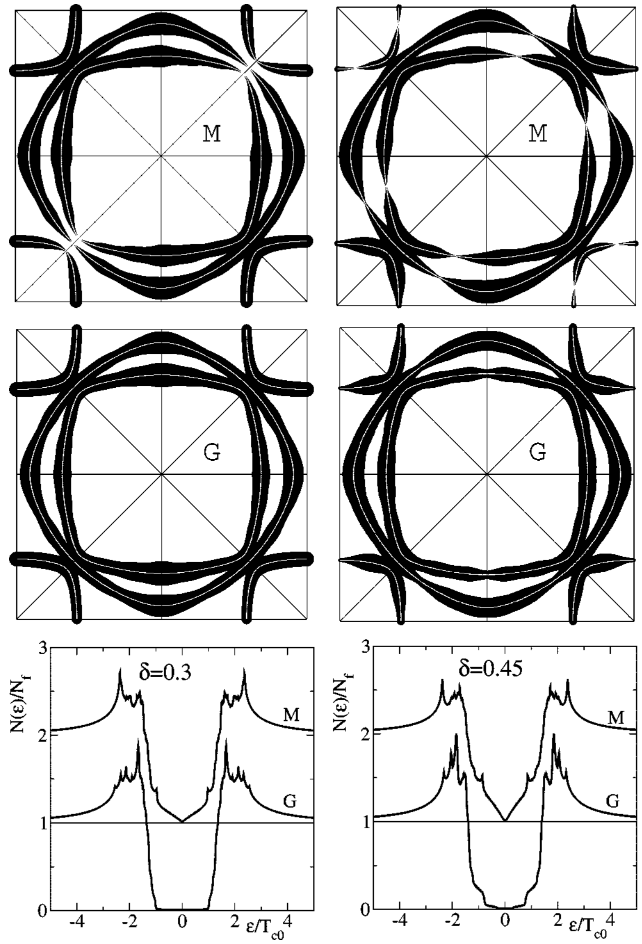

FIG. 3. The same as in Fig. 2 for $\delta=0.3$ (left) and for $\delta=0.45$ (right). Top to bottom: gap magnitude of the metastable state (M), gap magnitude of the ground state (G), and the density of states for each corresponding state.

Going to a δ where superconductivity nucleates first in either the B_{1g} or the A_{1g} channel, we find that already the ground states are mixed parity states. The lowest-energy state is a nodeless time-reversal symmetry-breaking state with relative phases similar to the $M1$ state discussed above.

For both $\delta=0.3$ and $\delta=0.45$ we find metastable states, with nodes on all three Fermi surface sheets, that are of slightly higher free energies (9.0 and 15.6 %, respectively) than the nodeless ground states. In Fig. 3 we show $|\Delta_+(\boldsymbol{p}_f)|$ and the DOS for both states at $\delta=0.3$ and $\delta=0.45$. For the metastable states the odd-parity components can develop higher order nodes, and remarkably, even whole arcs with vanishing or very small order parameter magnitude at the positions of the nodes in the even-parity components. At $\delta=0.3$, for instance, the odd parity component has nodes along a diagonal in the Brillouin zone, coinciding with the d-wave nodes of the even parity component. In this case such arcs of almost zero $\Delta^{(o)}(\boldsymbol{p}_f)$ occur on the γ sheet, extending all over the two quadrants which contain the nodes of the total order parameter.

The two components of the odd-parity order parameter transform like a vector in momentum space. Its direction is determined by the anisotropy introduced by (a) the normal-state DOS and (b) the presence of the even-parity B_{1g} or A_{1g} components. The mixed parity solutions for each incommensurability correspond to the alignment of this vector with high symmetry directions in the Brillouin zone. Because the energy difference between the ground-state and the state with nodes is only a fraction of the ground-state condensation energy, fluctuations in the direction of this vector can be responsible for the presence of nodal excitations. Analyzing data of specific heat measurements[5] and of thermal conductivity,[6] prompts that the Sr_2RuO_4 order parameter should have nodes.[8] This conclusion is further strengthened by recent measurements of the penetration depth showing a non-exponential low-temperature behavior.[7] Based on the presented calculations, this implies that a mixed parity superconducting state, breaking spin-rotation symmetry, may be a candidate state for Sr_2RuO_4, and may be even stabilized by additional interactions in the Hamiltonian not considered here.

In conclusion, we have shown that in the parameter region supported by experimental results even parity singlet and odd parity triplet pairing compete to have the highest transition temperature. Given the spin-spin correlation length of several lattice constants, as suggested by experiment, for an incommensuration near $\delta=0.35$ superconductivity nucleates in an accidentally degenerate $B_{1g}\oplus E_{2u}$ state and near $\delta=0.42$ the state is $A_{1g}\oplus E_{2u}$. Both states are of mixed parity and break the D_{4h} crystal symmetry and time-reversal symmetry. Within our model we find as ground states nodeless states which break time reversal symmetry. However, close in free energy there exist metastable states of the order parameter with nodes, which may be stabilized by additional interactions.

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